

Differential Photoelectron Holography: A New Approach for Three-Dimensional Atomic Imaging

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INTRODUCTION

Szöke [1] first suggested that highly coherent outgoing waves from atomically localized sources of photoelectrons, fluorescent x-rays, and γ -rays could be used to achieve atomic-scale holography. Among experimental approaches to such atomic-resolution holography, photoelectron holography (PH) has the advantages of being capable of studying the local atomic structure around each type of emitter without requiring long-range order and of distinguishing emitters through core-level binding-energy shifts [2]. However, atomic images in PH can suffer from serious aberrations due to the strong non-optical nature of electron scattering.

To overcome the difficulties associated with strong and anisotropic forward scattering (FS), we propose "differential holography". By simply replacing the normalized hologram $\chi(\mathbf{k})$ based on intensities measured over three-dimensional \mathbf{k} space by its k -derivative (i.e. $W = \partial/\partial k$) or more conveniently by a numerical difference between two χ 's at different energies ($\delta\chi = \chi(k + \delta k) - \chi(k)$), FS effects can be greatly suppressed [3]. We have applied this method to both experimental and theoretical multi-energy holograms for Cu 3p emission from Cu(001), and show that this provides images that are improved over prior work in several respects [4,5].

EXPERIMENTAL

To demonstrate differential PH (DPH) experimentally, photoelectron holograms from Cu(001) were measured at undulator beamline 7.0 of the Advanced Light Source at the Lawrence Berkeley National Laboratory. Photoelectron spectra for Cu 3p emission were collected at 25 energies for $k=4.5$ - 9.3 \AA^{-1} ($E_k=77$ - 330 eV) with a constant step of $\delta k=0.2 \text{ \AA}^{-1}$ ($\delta E_k=7.0$ - 14.0 eV) along 65 different directions over a symmetry-reduced $1/8$ of the total solid angle above the specimen and with a polar angle range from $\theta=0^\circ$ (surface normal) to 70° . A total of 1625 unique intensities were thus measured.

RESULTS AND DISCUSSIONS

Figures 1(a) and (b) show atomic images reconstructed from PH and DPH in the vertical (100) plane of Cu(001), respectively. In normal PH, only elongated features related to FS effects from atoms 6 and 7 are observed above $z_c=-0.5 \text{ \AA}$ (the arbitrary location of a change in image multiplication). Below z_c and with higher image amplification, several peaks near the back-scattering (BS) atomic positions 1-3 are observable among various strong artifacts, but only with

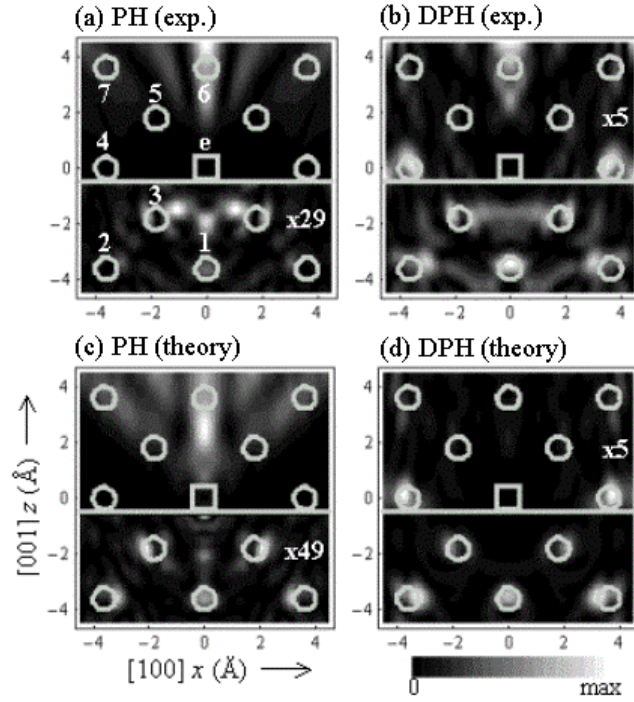


Fig. 1 Atomic images in the vertical (100) plane of Cu (001) reconstructed from Cu 3p holograms obtained by PH and DPH. The emitter and scatterer positions are indicated by squares and circles, respectively, and various near-neighbor atoms are numbered. Image intensities above or below $z_c = -0.5$ Å have been rescaled by the factor shown in each panel, with this factor being determined so as to make the maximum intensities above and below z_c equal. *Experimental images* (a) Image obtained by normal holography. (b) Image obtained by differential holography. *Theoretical images* (c) As (a) but theoretical. (d) As (b) but theoretical.

the help of a large scale factor of 29. In DPH, by contrast, the BS region is stronger than the FS region and the scale factor remains smaller at 5. A strong, somewhat elongated peak is observed at the FS atomic position 6, with weaker features that appear to be associated with atoms 7 also present in the corners of the image. Two strong peaks are observed at the side-scattering (SS) positions 4 above z_c . Also, the five strongest peaks below z_c are of roughly equal intensity and correspond reasonably well to the near-neighbor BS atoms. Therefore, we find DPH to be more robust than PH for imaging both SS and BS atoms (as well as to some degree FS atoms of type 6).

For comparison with experiment, we have also performed multiple-scattering simulations using a cluster method fully described elsewhere [6]. Images reconstructed from the theoretical χ and $\delta\chi$ are shown in Figs. 1(c) and (d), respectively. The main features in Figs. 1(a) and (b) are well reproduced by our simulations, although the artifacts between the images of atoms 3 are much stronger in experiment for PH, and the relative intensity in the region of FS atom 6 is stronger in experiment for DPH.

Finally, the full three-dimensional atomic image from the experimental $\delta\chi$ is shown in Fig. 2. We find in addition to the atoms of types 1-4 and 6 in Fig. 1, two other types of near-neighbor BS and SS atoms located in the vertical (110) plane (denoted as types 2' and 4' and situated in the same horizontal layers as 2 and 4, respectively). All of these atoms are reasonably well reconstructed, with only a few, such as 2, being significantly shifted in position, but most within a few tenths of an Å of the correct positions in all directions. Even though four weaker artifacts are observed at radii inside of the positions of atoms 3, the three-dimensional image quality is much better than any of the previous PH images of bulk substrate emission. The FS atom 6 is elongated as expected, but nonetheless has its brightest point rather close to the true location.

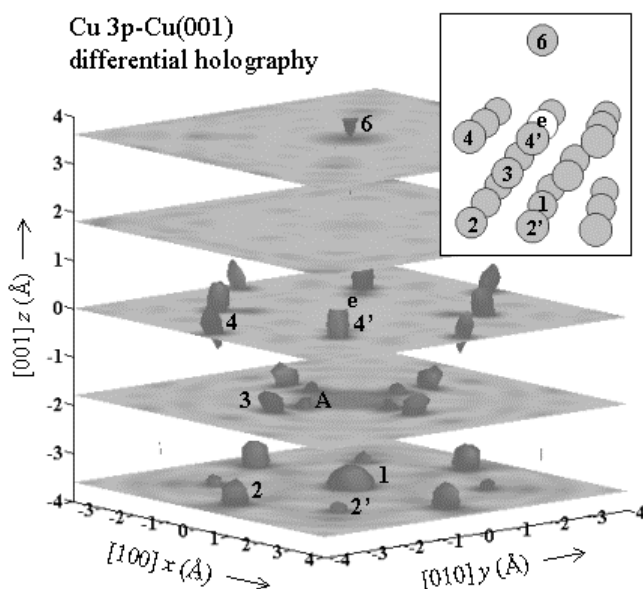


Fig. 2 Three-dimensional Cu(001) atomic image reconstructed from the experimental holograms by DPH. Isosurfaces at 50 % of the maximum intensity are shown together with five slices at $z=0$ (the emitter plane), ± 1.81 (the first layers of nearest neighbors) and ± 3.62 Å (the second layers). Here the scale factor for image intensities above $z_c = -0.5$ Å is 4. The origin is the emitter position “e”, and the ideal positions of the imaged atoms are indicated in the inset.

In summary, we have demonstrated differential photoelectron holography (DPH) as a powerful method for overcoming the FS problem in PH and enhancing image quality for any kind of system in which FS can arise, as for example, bulk emission, buried interfaces and complex overlayers. This method should also be helpful in other types of electron holography in which energy can be stepped in a controlled way (e.g. Kikuchi or LEED holography).

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